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Radiative transfer in clouds with small-scale inhomogeneities: Microphysical approach

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[1] Several recent publications have indicated that cloud droplets belonging to a particular size range may tend to cluster by forming small random particle groups imbedded in an otherwise uniform cloud. To analyze radiative transfer in clouds with such small-scale inhomogeneities, we invoke the concepts of statistical electromagnetics. We show that as long as the assumptions of ergodicity and spatial uniformity hold, one can still apply the classical radiative transfer equation in which the participating extinction and phase matrices are obtained by averaging the respective single-particle matrices over all the particles constituting the cloud. This result implies that comparisons of in situ and remote-sensing retrievals of the cloud-particle size distribution can be problematic and should be performed with caution. **Citation:** Mishchenko, M. I. (2006), Radiative transfer in clouds with small-scale inhomogeneities: Microphysical approach, *Geophys. Res. Lett.*, *33*, L14820, doi:10.1029/2006GL026312.

1. Introduction

[2] The microphysical approach to radiative transfer in particulate media was pioneered by *Borovoi* [1966] and has ultimately led to a self-consistent derivation of the general radiative transfer equation (RTE) directly from the Maxwell equations [*Mishchenko*, 2002, 2003]. This derivation has demonstrated that the RTE rests explicitly on the assumption that the scattering medium is statistically homogeneous. However, it has been suggested in recent publications [e.g., *Knyazikhin et al.*, 2002, 2005; *Shaw et al.*, 2002; *Marshak et al.*, 2005] that cloud droplets belonging to a particular size range may tend to form small groups of spatially correlated particles (clusters) imbedded in an otherwise homogeneous cloud.

[3] The problem of multiple scattering in stochastic particulate media has been studied so far using the concepts of the phenomenological radiative transfer theory [e.g., *Cairns et al.*, 2000; *Kostinski*, 2001; *Petty*, 2002; *Barker et al.*, 2003; *Davis*, 2006, and references therein]. However, this approach often implies the introduction of heuristic quantities with poorly defined physical meaning and numerous a priori assumptions not linked directly to a fundamental physical theory such as classical or quantum electrodynamics [see *Mishchenko*, 2006]. Hence the objective of this Letter is to address the problem of radiative transfer in particulate media with small-scale inhomogeneities by using the self-consistent microphysical approach explicitly based on the concepts of statistical electromagnetics.

2. Model, Assumptions, and Approximations

[4] We will use the following simplified model of an inhomogeneous cloud (Figure 1). The total volume of the cloud is V. The interior of the cloud is filled with $N_{\mathrm{bp}} \geq 0$ uniformly distributed "background" particles and $N_{\mathrm{i}} \gg 1$ uniformly distributed, small, compact inhomogeneities. Each inhomogeneity occupies a (nearly spherical) volume V_{i} such that

$$V_{\mathrm{i}} \ll V \quad \text{and} \quad N_{\mathrm{i}} V_{\mathrm{i}} < V \tag{1}$$

and is filled with a small number N_{ip} of "inclusion" particles such that

$$N = N_{\mathrm{bp}} + N_{\mathrm{i}} N_{\mathrm{ip}} \gg N_{\mathrm{ip}}, \tag{2}$$

where N is the total number of particles in the cloud. The average single-scattering and absorption properties of the background and inclusion particles are, in general, different. The background particles can, in principle, be absent (i.e., N_{bp} can be zero).

[5] The cloud is illuminated by a plane electromagnetic wave. The case of illumination by quasi-monochromatic (e.g., solar) light will be discussed later. Our primary objective is to predict the response of a (polarization-sensitive) detector of electromagnetic energy located at an observation point, either internal (observation point 1 in Figure 1) or external. The location of the external observation point is arbitrary and can, for example, be below (observation point 2) or above (observation point 3) the cloud.

[6] In the framework of the microphysical approach to radiative transfer [*Mishchenko*, 2002, 2003], we will make the following fundamental assumptions.

[7] 1. Each cloud particle, either background or inclusion, is located in the far-field zones of all the other particles.

[8] 2. The observation point is also located in the far-field zones of all the particles forming the cloud.

[9] 3. All scattering paths going through a particle two or more times are neglected (the Twersky approximation). Doing this is justified since $N \gg 1$.

[10] 4. The cloud is an ergodic scattering system so that averaging over time can be replaced by averaging over particle positions and states.

[11] 5. The position and state of each background particle are statistically independent of each other and of those of all the other background particles. The spatial distribution of the background particles throughout the cloud is random and statistically uniform.

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[12] 6. The position of each inclusion within the cloud interior is random and statistically uniform and independent of those of all the other inclusions.

[13] 7. The spatial distribution and states of the inclusion particles within each inclusion are independent of the position of this inclusion.

[14] 8. The position and state of each inclusion particle are statistically independent of each other. The state of each inclusion particle is independent of the positions and states of all the other inclusion particles belonging to the same inclusion. The spatial distribution of the inclusion particles throughout each inclusion is random and statistically uniform.

[15] 9. All diagrams with crossing connectors in the diagrammatic expansion of the coherency dyadic can be neglected (the ladder approximation).

[16] Note that the state of a particle collectively represents its microphysical characteristics such as size, refractive index, shape, orientation, etc.

3. Derivation of the Radiative Transfer Equation

[17] Assumptions 1–3 from the preceding section allow us to write the instantaneous electric field at the observation point in the form of the far-field order-of-scattering Twersky expansion:

$$
\begin{aligned}
\mathbf{E} \approx \mathbf{E}^{\mathrm{inc}} &+ \sum_{i=1}^{N} \overleftrightarrow{B}_{ri0} \cdot \mathbf{E}_i^{\mathrm{inc}} + \sum_{i=1}^{N} \sum_{\substack{j=1 \\ j\neq i}}^{N} \overleftrightarrow{B}_{rij} \cdot \overleftrightarrow{B}_{ij0} \cdot \mathbf{E}_j^{\mathrm{inc}} \\
&+ \sum_{i=1}^{N} \sum_{\substack{j=1 \\ j\neq i}}^{N} \sum_{\substack{l=1 \\ l\neq i \\ l\neq j}}^{N} \overleftrightarrow{B}_{rij} \cdot \overleftrightarrow{B}_{ijl} \cdot \overleftrightarrow{B}_{jl0} \cdot \mathbf{E}_l^{\mathrm{inc}} \\
&+ \sum_{i=1}^{N} \sum_{\substack{j=1 \\ j\neq i}}^{N} \sum_{\substack{l=1 \\ l\neq i \\ l\neq j}}^{N} \sum_{\substack{m=1 \\ m\neq i \\ m\neq j \\ m\neq l}}^{N} \overleftrightarrow{B}_{rij} \cdot \overleftrightarrow{B}_{ijl} \cdot \overleftrightarrow{B}_{jlm} \cdot \overleftrightarrow{B}_{lm0} \cdot \mathbf{E}_m^{\mathrm{inc}} + \cdots, \qquad (3)
\end{aligned}
$$

where the notation follows that in Section 3C of *Mishchenko* [2002]. In particular, $\mathbf{E}^{\mathrm{inc}}$ is the incident field, the indices i, j, etc. number the particles, and the scattering dyadics $\overleftrightarrow{B}$ describe the transformation of the electric field upon single scattering by a particle. The second term on the right-hand side of equation (3) describes the cumulative contribution of all scattering paths going through one particle, the third term represents the contribution of all double-scattering paths, etc. In accordance with the Twersky approximation, no scattering path is allowed to go through a particle more than once.

[18] To compute an actual optical observable, one must substitute the expansion (3) in the corresponding formula defining the observable and take an average over all realizable particle positions and states. The difference of the situation analyzed in this paper from that studied by *Mishchenko* [2002, 2003] is that now the positions of the inclusion particles belonging to the same inclusion are partly correlated. Indeed, although the position of an inclusion particle within an inclusion can be arbitrary, the distance from the center of the inclusion to the particle can never exceed the inclusion radius. This implies that the distance between any two inclusion particles can never exceed the inclusion diameter.

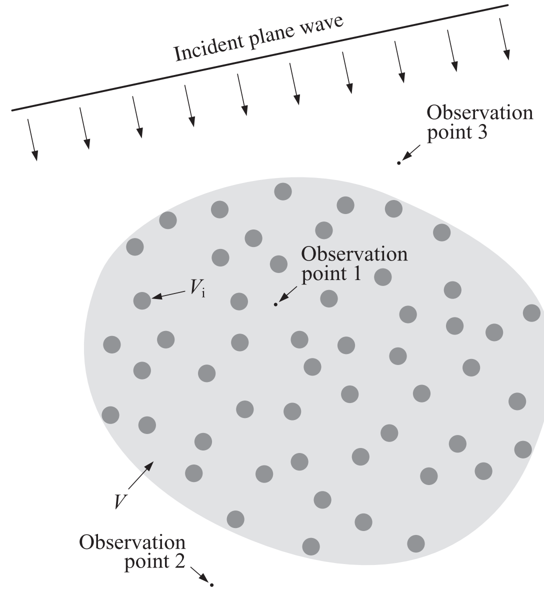

Figure 1. Cloud morphology.

[19] It follows from the Twersky expansion that this partial correlation affects only the scattering paths of the second and higher orders and only those going through an inclusion more than once. Indeed, statistical averaging of the second term on the right-hand side of equation (3) requires only the knowledge of the probability to find particle i inside an elementary volume dV centered at a point $\mathbf{r}$ within the cloud without regard to where the rest of the particles are located. For a background particle, the corresponding probability density function is simply $1/V$. For an inclusion particle, the corresponding probability is equal to the probability that the point $\mathbf{r}$ is inside the inclusion volume times the probability that the inclusion particle is located inside the elementary volume dV centered at the point $\mathbf{r}$, that is, $(V_\mathrm{i}/V)(1/V_\mathrm{i})\mathrm{d}V$, where we have taken into account the first inequality of equation (1). The resulting probability density function is again $1/V$.

[20] The direct evaluation of the effect of the partial spatial correlation of particles belonging to the same inclusion on the contributions of the multiple-scattering paths is an extremely difficult analytical problem. It can be shown, however, that this effect can be neglected in the case of small inhomogeneities owing to the inequality (2). Indeed, let us consider the contributions of the second-order scattering paths described by the third term on the right-hand side of equation (3). The total number of such paths is equal to $N(N-1)$, whereas the number of such paths that go through the same inhomogeneity twice and are, thus, affected by the spatial correlation is only $N_\mathrm{i} \times N_\mathrm{ip}(N_\mathrm{ip}-1) \ll N(N-1)$. Thus the contribution of the latter paths to the left-hand side of equation (3) can be neglected in comparison with the cumulative second-order contribution.

[21] This result can be reformulated by stating that the cumulative second-order-scattering interaction of a member of a cluster with its own cluster is much weaker than with the rest of the cloud. Indeed, a member of a cluster is involved in $N_\mathrm{ip}-1$ second-order scattering paths going through the other members of the same cluster and in $N - N_\mathrm{ip}$ second-order scattering paths going through all the other

particles forming the cloud. Admittedly, the scattering dyadic $\overleftrightarrow{B}_{ij0}$ includes a factor $1/R_{ij}$, where R_{ij} is the distance between particles i and j. The average value of this factor will be substantially smaller for two members of the same cluster then for a member of a cluster and any other particle forming the cloud. However, the inequality $N \gg N_{\mathrm{ip}}$ makes this circumstance insignificant, especially if one takes into account how large the total number of particles in a liquid-water cloud can be at the typical droplet number concentration $\sim 10^8$ particles/m^3.

[22] Indeed, let us consider, for example, a cluster consisting of $N_{\mathrm{ip}} = 10^3$ components and occupying a spherical volume with radius $R_{\mathrm{i}} = 10^{-2}$ m. The average inverse distance between a cluster component located in the center of the spherical volume and any other member of the cluster is proportional to $1/R_{\mathrm{i}} = 10^2$ m^{-1}. Let us now consider a spherical cloud volume with radius 10 m centered at the cluster. The average inverse distance between the central cluster component and any other particle belonging to the 10-m-radius cloud volume is proportional to 10^{-1} m^{-1}. However, the number of such cloud particles is $\sim 4 \times 10^{11}$. Obviously, the product $10^3 \times 10^2 = 10^5$ is much smaller than the product $10^{-1} \times 4 \times 10^{11} = 4 \times 10^{10}$ and becomes even less significant if one takes into consideration the entire cloud rather than only its 10-m-radius part.

[23] The reader can verify that the same is true of all higher-order scattering paths: the contribution of all the paths of an order ≥ 2 that go through the same inhomogeneity more than once can be neglected in comparison with the cumulative contribution of all the scattering paths of this order. This is a very important result which simplifies drastically all derivations. Indeed, since the former contribution is negligibly small anyway, one can evaluate it approximately by assuming that there are no spatial correlations at all. Then the entire derivation becomes exactly the same as in *Mishchenko* [2002, 2003] and yields exactly the same result.

[24] We can, thus, conclude that the transfer of radiation in clouds with small-scale inhomogeneities is adequately described by the classical radiative transfer equation. Furthermore, the corresponding extinction and phase matrices are obtained by the standard averaging of the respective single-particle matrices over all N particles constituting the cloud assuming that the background and the inclusion particles form a uniform particle mixture. This is the main result of this Letter. It is straightforward to show [cf. *Mishchenko et al.*, 2006, Section 8.15] that it remains valid in the case of illumination by quasi-monochromatic light, in particular, sunlight.

[25] One important implication of this result is that the classical exponential extinction law remains applicable to weakly inhomogeneous particulate media. The same conclusion was drawn previously by *Borovoi* [2002] on the basis of heuristic phenomenological considerations.

4. Discussion

[26] The applicability of the classical RTE to clouds with small-scale inhomogeneities is a quite welcome result since it permits the direct use of a number of well-known analytical and numerical solution techniques [e.g., *Lenoble*, 1985; *Hovenier et al.*, 2004]. Although it rests on several fundamental assumptions listed in Section 2, most of them appear to be quite plausible. However, the assumptions of ergodicity and spatial uniformity deserve a separate analysis.

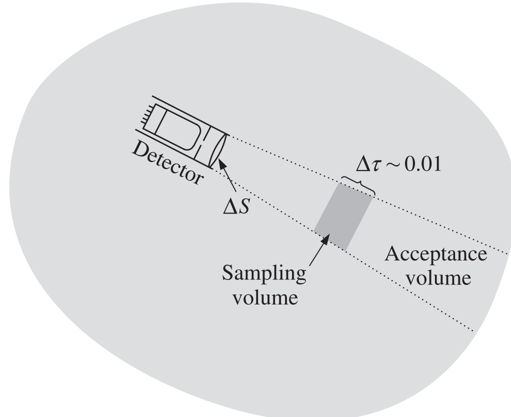

Figure 2. The practical meaning of the assumptions of ergodicity and spatial uniformity.

[27] The meaning of the assumptions or ergodicity [*Mishchenko*, 2006] and spatial uniformity is illustrated in Figure 2. The detector of electromagnetic energy has an angular aperture small enough to resolve the angular variability of the radiation field (e.g., $\sim 1°$) and a finite acceptance area ΔS. Both define the part of the cloud volume bounded schematically by the dotted straight lines in Figure 2; this volume will be called the acceptance volume. According to the integral form of the RTE, all energy recorded by the detector comes directly from the particles contained in the acceptance volume [cf. *Mishchenko*, 2002, equation (125)]. The energy exciting each particle can be either the (attenuated) sunlight or the light scattered by the other particles. The light scattered by a particle from the acceptance volume toward the detector can be attenuated by other particles located closer to the detector.

[28] Let us assume that the detector accumulates the signal over a time interval Δt and subdivide the acceptance volume into a number of sampling volumes such that their optical thickness $\Delta\tau$ along the line of sight of the detector is very small (~ 0.01). One of these sampling volumes is shown schematically in Figure 2. Obviously, the contribution of a particle to the detector signal is essentially independent of the specific particle position in the sampling volume. Therefore, the strict meaning of the assumptions of ergodicity and statistical uniformity of particle and inclusion positions within the cloud is that each particle visits each sampling volume during the measurement interval Δt.

[29] In reality, the cloud contains many particles of the same type. Therefore, the practical meaning of the assumptions of ergodicity and spatial uniformity is that particles of each type visit each sampling volume during the measurement interval Δt a number of times statistically representative of the total number of such particles in the entire cloud.

[30] It is quite reasonable to expect that the assumptions of ergodicity and spatial uniformity hold in passive satellite observations, in which case the large instantaneous geometrical field of view of a typical instrument (hundreds of meters or more) ensures a large size of each sampling volume. The same is true of the atmospheric radiation

budget computations, in which case the role of a detector of electromagnetic energy is played by large Earth surface areas. However, in situ measurements are often taken with relatively small instruments over short accumulation times. Therefore, one has to exercise care in comparing size distribution results obtained with remote-sensing and in situ detectors of scattered electromagnetic energy. The passive remote-sensing retrievals are likely to be representative of the size distribution averaged over the entire cloud, whereas the in situ measurements may depend strongly on the instrument characteristics such as Δt and ΔS and sometimes may be difficult to interpret in terms of a multiple-scattering theory. To ensure the validity of the assumptions of ergodicty and spatial uniformity in such cases one may need to increase Δt and/or ΔS quite significantly, which may or may not be practical.

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